

# *The Effect of Lead Antiknocks on the Lead Content of Crops*

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## *ABSTRACT*

Man receives on the average about 300  $\mu\text{g}$  lead/day in his food. This natural concentration of lead in food results from the lead present in the soil. The lead in soils averages about 16  $\mu\text{g/g}$  worldwide. Under conditions that have thus far been studied, lead in air does not measurably increase the lead content of the edible portion of most crops. Leafy portions of plants near busy highways contain higher concentrations of lead. Even in the absence of lead in air, leafy portions of plants contain more lead than do other parts. A several-fold increase in lead in soil does not measurably change the concentration of lead in plants. Rainwater does not appear to be a significant source of lead in crops. All studies to date indicate that the effect of lead antiknocks on lead in the food chain is minimal.

## *Lead in Food*

To understand the effects of the use of lead antiknocks on the lead content of plants, one must consider some basic facts on the concentrations of lead in food. Much work has been done on the concentrations of lead in food. Schroeder and his coworkers (1961) have probably made the most complete examination of lead in food. On a fresh-weight basis, they found about 1.2 mg Pb/g in condiments, 0.5  $\mu\text{g/g}$  in fish and seafood, 0.2  $\mu\text{g/g}$  in meats, 0.4  $\mu\text{g/g}$  in

grains, 0.2  $\mu\text{g/g}$  in vegetables, and no detectable lead in fresh whole milk. Assuming a person consumes about 2,000g of food and drink a day, his lead intake would range from 100-500  $\mu\text{g/day}$  depending on the foods eaten. Cholak and Bambach (1943) estimated the intake of lead from food to be about 300  $\mu\text{g/day}$ . Kehoe (1947) estimated a similar amount. Monier-Williams (1950), as well as Warren and Delavault (1962), estimated about 0.2  $\mu\text{g Pb/g}$  of food which, based on 2000g of food, would be a lead intake of 400  $\mu\text{g/day}$ .

Kehoe et al. (1933) found lead in every item of food obtained from the fields and dwellings of the inhabitants of a primitive area, completely removed from industrial and mining activities.

Harley (1970) conducted an especially useful study in New York City. He determined the lead concentration in various foods and estimated the yearly intake of lead from the U.S. Department of Agriculture consumption statistics for food. Table 1 shows these results. The total annual lead intake of 103 mg/year or about 285  $\mu\text{g/day}$  is consistent with the results of other investigations.

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In 1965, he began work under an industrial cooperation agreement between the Ethyl Corporation and the Argonne National Laboratory, involving the investigation and analysis of trace metals in the environment. This work has covered all environmental aspects of lead, including its ultimate fate in the environment. These investigations have resulted in a number of publications.

Table 1. — Lead in New York City diet — 1966 sampling.

Diet Category	Food Intake, kg/year	Lead Intake	
		mg/kg food	mg/year
Dairy Products	200	0.04	8
Fresh Vegetables	48	0.12	6
Canned Vegetables	22	0.44	10
Root Vegetables	10	0.07	1
Potatoes	38	0.17	6
Dried Beans	3	0.02	—
Fresh Fruit	59	0.07	4
Canned Fruit	11	0.25	3
Fruit Juices	28	0.09	3
Bakery Products	44	0.39	17
Flour	34	0.04	1
Whole Grain Products	11	0.13	1
Macaroni	3	0.08	—
Rice	3	0.04	—
Meat	79	0.42	33
Poultry	20	0.30	6
Eggs	15	0.22	3
Fresh Fish	8	0.16	1
Shellfish	1	0.31	—
Annual Intake			103

Lewis (1966) reported that no food or group of foods is either a large or constant contributor to lead in man, since man's diet is composed of a wide variety of individual items, and various foods contributed various amounts of lead. Lewis estimated that the lead intake from the diet averages about 300  $\mu\text{g/day}$  and ranges, for most people, between 100 and 200  $\mu\text{g/day}$ . It appears that the average lead intake from food has not changed appreciably during the past 3 decades.

Although the lead intake from food averages about 300  $\mu\text{g/day}$ , the lead intake could vary markedly from city to city. Economic level and ethnic background also could have a pronounced effect on the amount of lead ingested daily. No studies attempting to answer this question have been reported. If such a study were carried out, it would be informative to determine the degree of correlation between the concentration of lead in the blood and the lead in food.

With lead intake from food in the human diet established to be of the order of 100 to 2000  $\mu\text{g/day}$ , the question arises as to the source of this lead. Patterson (1965) estimated that the natural lead content of food

Table 2. — Relative U.S. consumption of crops studied.

Crop	% Diet
Leaf Lettuce	0.5
Carrots	0.5
Head Cabbage	0.7
Snap Beans	1.0
Tomatoes	2.0
Sweet Corn	2.1
Potatoes	5.6
Wheat	10.9

should be 0.01  $\mu\text{g/g}$  wet weight. He concluded that most of the lead now present in the food is from industrial sources.

Effects of Lead  
in Air, Water, and Soil

To determine whether this latter claim is true, experiments have been designed to answer the following questions:

- What is the contribution of lead naturally present in the soil to the lead content of plants?
- What effect does lead in air have on this natural lead content of plants?
- What is the effect on the lead content of the plant from increasing the lead content of the soil from lead deposited from the atmosphere or from lead added artificially.

We attempted to answer the first question by growing crops in greenhouses using filtered air. We then compared the lead concentrations in these crops with those grown in unfiltered air to answer the second question. The crops chosen for this study and their relative contribution to the diet are shown in Table 2.

The soil used in the greenhouses was chosen because it was likely it had not been contaminated with industrial lead. It contained 17.1  $\mu\text{g Pb/g}$  dry, very near the world average of 16  $\mu\text{g/g}$  dry (Swaine, 1955). The unfiltered air contained 1.45  $\mu\text{g Pb/m}^3$  and the filtered air contained 0.09  $\mu\text{g Pb/m}^3$ .

The crops grown in the greenhouse were tomatoes, sweet corn, leaf lettuce, head cabbage, snap beans, potatoes, carrots, and wheat. All crops were harvested at maturity, washed, dried, dry ashed and brought into solution for analysis. Lead concentrations

Table 3. — Lead content of greenhouse crops.

Crop	Lead Content, $\mu\text{g/g}$ dry weight	
	Unfiltered Air	Filtered Air
<b>Edibles</b>		
Leaf Lettuce	<u>6.6</u>	3.2
Cabbage Head	<u>1.0</u>	1.1
Tomatoes	<u>0.6</u>	0.7
Snap Beans	<u>1.4</u>	1.2
Sweet Corn	<u>0.2</u>	0.3
Carrots	<u>1.7</u>	2.1
Potatoes	<u>0.3</u>	0.3
Wheat	<u>0.18</u>	0.16
<b>Nonedibles</b>		
Bean Leaves	<u>20.9</u>	7.9
Corn Cobs	<u>0.5</u>	0.7
Cabbage Leaves	<u>4.5</u>	5.8
Corn Husks	<u>6.9</u>	1.8

Underlined values are different from the other numbers in the row at the 95% level of confidence.

were determined colorimetrically using dithi-zone (Association of Official Agricultural Chemists, 1965).

The results of the greenhouse studies are summarized in Table 3. All data were handled by standard statistical methods using analysis of variance (Uni. Calif., 1966).

The data from this study answer our first question and show that the concentration of lead in the edible portion of the plants grown in filtered air ranges from a few tenths of a  $\mu\text{g}$  to a few  $\mu\text{g/g}$  of dried material. When the concentration is calculated on a wet-weight basis, the concentration is a few tenths of a  $\mu\text{g/g}$  for each of the crops. This concentration is of the same magnitude as that for foods purchased in the market place and is derived from the lead naturally present in the soil.

When we compare the concentrations of lead in the crops grown in filtered air with those grown in unfiltered air, we have some information that will answer the second question on effect of lead in air. All of the edible portions of the plants, except leaf lettuce, showed no effect from increasing the lead content of the air.

Although crops with a low exposed surface-to-weight ratio showed no effect of lead in air, plant parts having a relatively large exposed surface-to-weight ratio, primarily inedible, contained more lead when grown in unfiltered air than in filtered air. Thus, leaf lettuce, bean leaves, and corn husks showed an effect of lead in air.

We can obtain additional information with regard to the lead-in-air effect and some information with regard to the third question posed above on the lead-in-soil effect by studying crops grown in long rows perpendicular to a busy highway.

The same crops studied in the greenhouses plus oats and soybeans were grown in long rows perpendicular to and east of a heavily traveled north-south highway (U.S. 24 near Detroit, Mich.) with a traffic density of 29,000 cars/day.

Crops were harvested 30 to 60, 120, and 520 ft from the edge of the paved surface of the road. The average concentration of lead in the air during the growing season was 2.3, 1.7, and 1.1  $\mu\text{g}/\text{m}^3$  at 50, 120, and 520 ft from the road. The concentration of lead in the soil averaged 65  $\mu\text{g/g}$  at 40 ft from the road, 40  $\mu\text{g/g}$  at 120 ft, and 25  $\mu\text{g/g}$  at 520 ft.

In a similar study, samples of commercially grown rice were taken at 30 and 700 ft from U.S. Highway 90 (5,000 to 7,000 cars/day) just north of Crowley, La., and 45 and 600 ft from Interstate 10 (7,500 to 10,000 cars/day) just west of Crowley. At U.S. 90, the concentration of lead in the soil was 22  $\mu\text{g/g}$  at 30 ft from the road and 18  $\mu\text{g/g}$  at 700 ft. At Interstate 10, the lead in soil was 18  $\mu\text{g/g}$  at 45 ft and 15  $\mu\text{g/g}$  at 600 ft. The rice was hulled, and the kernel was analyzed for lead.

Table 4 shows the results of these studies. The results are generally consistent with those obtained in the greenhouse study. Edible portions of most compact crops (i.e., cabbage, potatoes, sweet corn, tomatoes, oats, wheat, carrots, and rice) showed no correlation between lead concentration and distance from the road. This implies that neither increasing the lead in the air from 1.1 to 2.3  $\mu\text{g}/\text{m}^3$  nor increasing the lead concentration in the soil by airborne deposition from 25 to 65  $\mu\text{g/g}$  had any effect on the edible portions of these plants.

In contrast to the greenhouse crops, however, 2 compact crops—soybeans and snap beans—showed higher lead concentrations when grown near the road. The reason for this inconsistency between the greenhouses and the roadside plots is not obvious.



Table 4. — Highway studies.

	Lead Content at Feet From Road		
	30	120	520
Air, $\mu\text{g Pb/m}^3$	2.3	1.7	1.1
Soil, $\mu\text{g Pb/g}$	<u>65</u>	<u>40</u>	<u>25</u>
Edibles, $\mu\text{g Pb/g dry}$			
Leaf Lettuce	6.5	5.0	4.5
Cabbage Head	0.56	0.86	0.83
Tomatoes	1.3	1.2	1.6
Snap Beans	<u>1.9</u>	1.2	0.9
Potatoes	0.48	0.64	0.40
Sweet Corn	0.39	0.21	0.83
Carrots	1.6	—	1.5
Soy Beans	<u>0.28</u>	0.12	0.10
Oats	0.47	—	0.37
Wheat	0.62	0.42	0.48
Rice (U.S. 90)	0.17	—	0.18
Rice (I-10)	0.23	—	0.24
Nonedibles, $\mu\text{g Pb/g dry}$			
Cabbage (unharvested leaves)	<u>6.4</u>	<u>8.9</u>	4.0
Corn Cob	0.74	0.55	0.68
Corn Husk	12.6	6.6	5.7
Soybean Husk	<u>15.9</u>	8.0	5.3
Oat Chaff	<u>31.4</u>	15.5	12.8
Wheat Chaff	<u>17.8</u>	<u>9.8</u>	6.2
Rice Straw (U.S. 90)	<u>4.1</u>	—	2.5
Rice Straw (I-10)	<u>5.83</u>	—	2.13

Underlined values are different from the other numbers in the row at the 95% level of confidence.

The pH of the soil could be a factor, although the variation was only about one pH unit. The pH of the plots along the highway varied from 7 at 30 ft from the road to 6.6 at 120 ft and 5.9 at 520 ft, while the pH of the greenhouse soil was 7.2.

The inedible parts of the plants (i.e., corn husks, wheat chaff, rice chaff, oat chaff, soybean hulls, and the broad normally unharvested outer leaves of cabbage) contained 2-3 times higher concentrations of lead when grown near the road compared to farther away.

Dedolph et al. (1970) conducted a similar study on grass and radishes. They studied the effects of lead in air, water, and soil on the concentration of lead in these 2 crops. They found that varying the concentration of lead in water from 1 to 40  $\mu\text{g/l}$  had no effect on the concentration of lead in these crops whether the water was applied to the foliage or to the surface of the soil.

They found no effect of lead-in-air concentration on radishes, but the grass was affected by lead in air. Both grass and radishes were found to derive about 2-3  $\mu\text{g}$

Pb/g dry matter from the soil when the concentration of lead in air was nil. When the concentration of lead in air was increased to about 1  $\mu\text{g/m}^3$ , the concentration of lead in the grass was about doubled.

Studies near a busy highway confirmed these results. Grass near the road contained about twice as much lead as grass grown 120 and 520 ft from the road. They concluded that plants contain substantial amounts of soil-derived lead and that soil has long been and remains an important source of lead in plants.

Many authors substantiate the conclusion that lead in air does increase the concentration of lead in the leafy parts of plants near the highway. Koke and Riebartsch (1964) found higher concentrations of lead in grass grown near busy highways. Cannon and Bowles (1967) found higher concentrations of lead in vegetation grown near a highway than in vegetation grown some distances away. Warren and Delavault (1962) compared lead in plants to highway traffic. They determined the lead content of tree stems collected in an area at locations remote from the highway and adjacent to heavy traffic. The lead values ranged from 0.4 to 2.0  $\mu\text{g/g}$  dry for the "remote" stems and 2-52  $\mu\text{g/g}$  dry for the "heavy traffic" stems. Everett et al. (1967) measured the lead content of unwashed privet leaves collected from sites along main highways and remote from highways in England. They found an average of 86  $\mu\text{g Pb/g}$  dry in the leaves near the highway and 45  $\mu\text{g Pb/g}$  dry at the sites away from the highways.

All investigators reach the same conclusion. In a narrow band near the highway, the concentration of lead on the surface of foliage is proportional to the concentration of lead in air. On the protected portions of the plants (e.g., seeds and roots), which in almost all cases are the edible portions of the plants, little or no effect of lead in air is noted.

Even in the absence of lead in air, the leafy portions of the plants are higher in lead than the rest of the plant. Ter Haar (1970) observed this in his greenhouse study. Goldschmidt (1937) observed this as early as 1937. He stated that the mineral solution

enters the plants through the roots and concentrates at the point of greatest evaporation, namely the leaves.

Studies by Motto et al. (1970) on the effects of lead in air and soil indicate similar results. They found that the major effect of traffic was limited to a narrow zone within 100 ft of the highway. Plants grown in the field contained the most lead in the aerial portions. They found that lead was absorbed through the root system with some translocation to other parts of the plant. The fruiting and flowering parts of the plant contained the smallest amount of lead and showed little effect of changes in the amount of lead supplied.

Leh (1966) also observed that higher concentrations of lead were found in vegetation near a highway. He found higher concentrations of lead in grass, turnip and beet leaves, and chaff from wheat and barley grown near an expressway. However, he found no effect of lead in air on potatoes, beets, turnips, carrots, celery, or the grain kernel.

Schuck and Locke (1970) studied 5 crops — cauliflower, tomatoes, cabbage, strawberries, and oranges. They reported that the combined findings on the edible portions from 4 of these 5 crops strongly suggest that automotive lead particulates are not absorbed, but rather exist as a topical coating of which at least 50% can be removed by simple water washing. In the case of the fifth crop (strawberries), washing did not remove lead from the fruit. The concentration of lead in the strawberries was not influenced by distance from the road. They also found that the crops did not show an inclination to absorb lead by the root system. In spite of growing these crops near heavily traveled highways with up to 50,000 cars/day, the amount of lead associated with the 5 crops in an untreated state was never greater than 1  $\mu\text{g}$  Pb/g fresh weight. The average lead concentration for the entire crop area studied was 1 or 2 orders of magnitude less than the 1  $\mu\text{g}$  Pb/g fresh weight.

Although their conclusion that crops are not inclined to absorb lead through the root system disagrees with the conclusions of

some of the authors previously cited, it may be that the pH of their soil or some other physical or chemical characteristic of the soil led to this conclusion. Lagerwerff (1970) stresses the importance of pH in lead uptake into plants. Little work has been done on the uptake of lead from different soil types. The effects of pH and other chemical variables also remain to be investigated.

In all of these studies, the lead in the soil was higher near the road. The crops take up lead from the soil in a relatively constant manner, which is independent of several-fold changes in the lead concentration of the soil. Marten and Hammond (1966) found that a 52-fold increase in the lead content of the soil taken near a smelter increased the lead content of brome grass when grown on the soil in a greenhouse in the first harvest. The second harvest did not contain significantly more lead when grown in soil containing 680  $\mu\text{g}$  Pb/g than grass grown in soil containing 12  $\mu\text{g}$  Pb/g. When grass was grown in a greenhouse on a soil taken near a busy highway (59  $\mu\text{g}$  Pb/g), the lead concentration of this grass was the same as that of grass grown on the soil containing 12  $\mu\text{g}$  Pb/g. Soil at a 15-cm depth taken near the smelter contained 95  $\mu\text{g}$  Pb/g. It also had no effect on the concentration of lead in the grass.

A study by MacLean et al. (1969) shows the importance of soil type, cation exchange capacity, carbon content, and crop type on the effects of the lead concentration in plants resulting from adding lead to the soil. They found that a soil with high exchange capacity was much less likely to release lead to the plant than a soil with low exchange capacity. The oat kernel was much less affected than the oat straw. Alfalfa was affected more than oat straw. The addition of phosphorus to the soil markedly reduced the uptake of lead.

#### Seasonal Variation

One final problem to be concerned about when studying the lead content of plants is discussed by Mitchell and Reith (1966). They found that the lead content of the whole above-ground portion of a plant in-

creases when active growth stops. The lead content of the above-ground portion of pasture herbage increased from about 1  $\mu\text{g/g}$  dry in the summer to 10  $\mu\text{g/g}$  dry in the autumn, and reached 30-40  $\mu\text{g/g}$  dry in the winter. The authors believe that the increase in lead content of the above-ground portion when the plant is dormant may indicate movement from the root rather than uptake from the soil. They rule out the possibility of surface contamination from lead in air as well as soil contamination.

This study indicates caution is necessary when comparing lead concentrations in materials. If they are harvested in different seasons, the results may not be comparable. More work is needed in this area of seasonal variation and the effect of stress on lead uptake.

#### Conclusions

- Man receives an average of about 300  $\mu\text{g}$  Pb/day in his food, but the range may be from 100 to 2000  $\mu\text{g}$  per day.
- The natural concentration of lead in food resulting from the natural lead content of the soil is a few tenths of a  $\mu\text{g/g}$  wet.
- Lead in air does not measurably increase the lead content of the edible portion of most plants.
- A several-fold increase in lead in the soil does not measurably change the concentration of lead in the plant.
- Leafy portions of plants near busy highways clearly contain higher concentrations of lead. This is true for a narrow band on both sides of the highway.
- Even in the absence of lead in air, leafy portions of plants contain more lead than do the other parts.
- Rainwater does not appear to be a significant source of lead in crops.
- Stresses on the plant, such as senescence, may increase the concentration of lead.

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## *The Effect of Outboard Motor Exhaust Wastes on Fish and Their Environment*

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### ABSTRACT

Bluegill sunfish were placed in liveboxes and sampled at two-week intervals in (1) a lake where much water skiing occurred, (2) in a pond where outboard motors with low-pitched propellers were operated by project personnel, and (3) in a control pond where outboard motors were not operated. The fish were fried in vegetable oil and cracker meal at a temperature of 370°F (188°C) or baked in aluminum foil before being tasted by a taste panel of 12 members. The tainting of fish occurred at a level of about 2.6 gal outboard motor fuel/acre-ft of water or 8 gal fuel/million gal water, and a daily fuel-use rate of 0.17 gal/million gal water (0.055 gal/acre-ft). Threshold odor, carbon chloroform extractables, and chlorine demand showed significant increases in the motor lake and motor pond through the season of outboard motor operation. All water samples from the motor lake and motor pond contained less than 10 µg/l of lead determined by polarograph.

Laboratory tests conducted in 1960 at the Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, by English et al. (1963a) showed that bluegill sunfish could be tainted by outboard motor exhaust wastes. Ninety % of persons in a taste panel noted objectionable flavor at a cumulative fuel consumption of 8.6 gallons per acre-foot of water. Half of the panel members

noted objectionable taste at 1.1 gallons of fuel per acre-foot.

These laboratory experiments did not show toxicity to fish until the fuel consumption reached 170 gal/acre-ft when half of the fish were killed in 96 hr at a dilution of about 1 in 5. The 96-hour TL<sub>m</sub> was, therefore, 19%. When an application factor of 10 was applied for the estimation of the "safe" level, this was projected to 17 gal fuel/acre-ft of water.

Complaints of off-flavoring in fish reached us from a relatively small Ohio lake which was surrounded by cottages and where there was intensive use of outboard motors. These complaints as well as the results of the studies by English et al. reported briefly above, emphasized the need for field studies

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